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(54) Title: GOLD-NICKEL-VANADIUM BRAZING MATERIALS (57) Abstract A ductile brazing material containing 75-98 % gold, 0-20 % nickel, 0.5-6 % vanadium and, optionally chromium and/or molybdenum at less than about 6 % is disclosed for directly bonding ceramic to ceramic or ceramic to metal over an optimum temperature range.		

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GOLD-NICKEL-VANADIUM BRAZING MATERIALS

This invention relates to active metal brazing materials. More particularly, the invention relates to brazing alloys and pastes for brazing metal to metal, metal to ceramic, or ceramic to ceramic.

Active metal brazing is a technique in which wetting and bonding of the braze material to ceramic or oxidised metal surfaces is improved due to the presence of a small amount of active metal such as titanium, vanadium, chromium or zirconium. Such brazing metals provide chemical as well as mechanical bonding to substrates. A review of such alloys may be found in 'Ceramics and Glasses', Volume 4, Engineered Materials Handbook pages 502-510 (published by ASM International, 1991). A further review may be found in 'Joining of Ceramics', pages 73-92, editor M.G. Nicholas, (published by Chapman & Hall for the Institute of Materials, 1990).

U.S. Patent No. 4,447,391 claims active brazing alloys comprising:-

0.1-5%	reactive metal (titanium, zirconium, vanadium or mixtures thereof)
0.05-4%	boron
15-85%	nickel
0-30%	chromium
5-85%	precious metal (gold, palladium, or mixtures thereof)

but only exemplifies compositions in which titanium is the active metal and indeed states that titanium is the "especially preferred" reactive metal.

U.S. Patent No. 4,606,978 claims active brazing alloys comprising:-

0.25-5%	titanium
3-40%	nickel
5-65%	copper
25-85%	gold

and optionally claims chromium in amounts 2-15%. This document also discloses but does not exemplify use of vanadium, zirconium, titanium or mixtures thereof as active metal in the same way as U.S. Patent No. 4,447,391 above.

U.S. Patent No. 4,938,922 claims active brazing alloys comprising:-

0.1-2	titanium
0.5-7%	nickel
91-99%	gold

U.S. Patent No. 3,846,125 claims gold alloy wires for use as potentiometer wires and the like comprising:-

7-8%	vanadium
5%	iron
balance	gold

but also states that nickel may be used in replacement for gold. Only one example is shown indicating use of nickel in replacement of iron (Sample code 659) and this is shown as having a composition 90% gold, 3% vanadium and 75% nickel (i.e. 168% in total!). Making the assumption most charitable to the applicant for that patent the composition might be 90% gold, 3% vanadium and 7.5% nickel (i.e. 100.5% in total).

U.S. Patent No. 4,820,487 claims decorative gold alloys in the form of alloy bars comprising:-

1-3%	vanadium
3-6%	nickel
2-3%	chromium
0.1-0.4%	molybdenum
0.1-1.5%	carbon
0.1-1.5%	tungsten
3-6%	iron
80-88%	gold

Japanese Patent Specification No. 63-317276 discloses a gold filler alloy for brazing Mo-Mn metallised surfaces and comprising:-

0.001-15%	titanium, zirconium, vanadium or niobium
10-27%	nickel
balance	gold

but the only examples disclosed which exemplify use of vanadium have a nickel content of greater than 15%.

Brazing to Mo-Mn metallised surfaces has the disadvantage of requiring extra metallisation steps in processing.

It is well known in the art of brazing that the thermal expansion mismatch between many materials, particularly a metal and ceramic member, requires the use of a ductile braze material. Thus, brittle brazing materials, such as the gold-based alloys disclosed in U.S. Patent No. 4,447,391 (containing boron), have limited usefulness.

However, the gold-nickel-titanium brazing alloys disclosed in U.S. Patent No. 4,938,922 are illustrative of ductile brazing materials. In order to achieve the desired ductility the titanium content of these alloys is generally maintained at very low levels, i.e. 0.1% to 2.0% weight percent. The disclosed gold-nickel-titanium alloys may be employed in a single step process which produces a highly ductile brazed joint with excellent oxidation resistance at 650°C and no visible reaction to acid and alkali treatment. The alloy has been successfully employed to braze silicon nitride ceramic to Incolloy 909 alloy for use in internal combustion engines, and is currently produced and sold under the trademark SNW-3000 by Wesgo Inc., a subsidiary of The Morgan Crucible Company plc.

The gold-nickel-titanium alloys of U.S. Patent No. 4,938,922 are also useful for brazing superalloys such as Inconel 718 and other superalloys containing aluminium and/or titanium. Both aluminium and titanium are strong oxygen getters resulting in a thin layer of oxide upon heating such superalloys, making wetting with conventional alloys difficult. Wetting can be improved by increasing the temperature but only to the detriment of prior brazes made at higher temperatures or the materials being brazed. For example, a 50% gold - 50% copper alloy with a liquidus temperature of 970°C will typically flow at about 1,000°C. However, when brazing an Inconel 718 alloy, the brazing is generally performed at about 1,070°C. A copper member with a melting point of 1,083°C may thus tend to melt close to a furnace heating element since most commercial furnaces exhibit temperature fluctuations of about 10-25°C within the furnace area.

The gold-nickel-titanium alloys of U.S. Patent No. 4,938,922 will exhibit sufficient wetting to an alumina ceramic, but, over a very narrow temperature range of about 20°C. Exceeding this temperature results in "dewetting" - that is, the molten alloy beads up leaving bare ceramic where molten alloy formerly coated the surface.

The applicants have found that if vanadium is substituted for titanium as the active metal higher quantities of vanadium may be employed while still maintaining the desired ductility. The increased quantity of active metal significantly enhances the wetting characteristics of the brazing material over a broader range of temperatures.

As a corollary to this it has been found that, for a given level of reactive metal, vanadium containing materials are more ductile than titanium containing materials. The brazing material will therefore exhibit a lower yield strength. The lower yield strength results in lower residual stress in a brazed joint since the plastic deformation of the brazing material accommodates the thermal expansion mismatch between articles being brazed. The brazing material will also be easier to mechanically reduce/deform (e.g., in rolling processes) so minimising any edge cracking.

The applicants have also found that for the higher gold compositions of the present invention (i.e. gold content greater than about 92% by weight) use of molybdenum may be tolerated up to e.g. 4%. This is in contrast to the alloys of U.S. Patent 4,606,978 which discloses use of molybdenum at 6% or more to prevent creep: prevention of creep at first sight would be thought to reduce ductility and so impair performance. For the lower gold compositions of the present invention (gold < about 92%) molybdenum is beneficial in small amounts (i.e. up to 5.9% preferably less than 4% by weight) in that it improves ductility.

Accordingly the present invention provides a method of forming a brazed joint between a first material and a second material, which method comprises immediately disposing between said first and second material a brazing composition comprising in weight percent (incidental materials and volatile constituents excepted):-

75-98% gold
0-20% nickel
0.5-6% vanadium
0-5.9% molybdenum
0-6% chromium.

The lower level for nickel is preferably 0.25% as this assists wetting.

In one aspect of the invention a higher gold brazing material has a composition substantially comprised of, in weight percent:

92-98% gold
0-7.5%, preferably 0.25-7.5%, more preferably 0.5-3% nickel
0.5-6% vanadium
0-4%, preferably 0-1% molybdenum
0-6%, preferably 0.3-3%, more preferably 1-3% chromium.

A preferred range of vanadium content is about 1.5-3.5% by weight vanadium, more preferably about 1.75% by weight such as 1.5-2.0%. Thus, in a preferred embodiment, the higher gold brazing material composition substantially comprises, in weight percent: 94.5-97.5% gold, 0.5%-3% nickel, 1.5-2.0% vanadium.

The present invention further provides higher gold gold-nickel-vanadium brazing materials comprising alloy foils or pastes that on melting will form a material of the claimed composition.

In a second aspect the invention also provides a method of brazing using lower gold content alloys (gold less than 92% by weight) comprising in weight percent (incidental materials and volatile constituents excepted):-

75-92% gold
0-20% nickel
0.5-6% vanadium
0.25-5.9%, preferably 0.5-4%, molybdenum
0-6%, preferably 0.3-3%, more preferably 1-3% chromium.

For this second aspect of the invention, when used to braze nickel-containing alloys, the nickel content of the

brazing composition is preferably kept in the range 14-16.5%. As the level of nickel is reduced below this the melting point increases and the erosive character of the brazing composition for nickel-containing alloys increases; both trends are undesirable. Accordingly in a preferred embodiment the gold content is less than 85%, preferably about 82%.

The invention further provides such alloys in the form of foils and pastes.

It has been found that the brazing materials of the present invention produce highly ductile materials which are particularly useful for brazing ceramics. The gold-nickel-vanadium brazing materials also exhibits excellent wetting to ceramics and does not dewet when exposed to a wider brazing temperature range as compared to a gold-nickel-titanium brazing materials. Thereby the materials of the present invention provide a brazing material that can be directly brazed to a ceramic surface over a broader range of temperature than titanium containing alloys and which yield a highly ductile, oxidation and corrosion resistant brazed joint.

The gold-nickel-vanadium brazing materials of the present invention generally have liquidus temperatures in the range of from about 900°C to about 1,100°C. The brazing materials are highly ductile and, after brazing, are relatively free of hard dispersed phases.

The lower gold materials of the present invention show a generally lower melting point than the higher gold materials of the present invention and this is useful when brazing metals as the alloys hence have little dissolving effect on metals. This is useful when brazing e.g. superalloys and refractory metals such as molybdenum, tungsten, niobium or platinum.

The compositions of the present invention allow direct brazing of surfaces in contrast to the method of Japanese Patent Specification No. 63-317276 which requires Mo-Mn metallisation.

According to a further aspect of the invention, chromium may be added to improve corrosion and oxidation resistance.

It has been found that the addition of chromium to both the higher and lower gold gold-nickel-vanadium brazing materials of the invention also results in an increase in ductility. Further, a low concentration of chromium (e.g., 0.5-1.5%) permits lowering of the vanadium content without affecting the "wettability" of the brazing material to ceramic surfaces.

Chromium is soluble in the higher gold alloys of the invention up to about 6% by weight. However high chromium levels of around 6% may raise both melting point and hardness which are not desirable features in a brazing alloy. Accordingly a preferred upper level for chromium (if present) is 3%. In the absence of chromium no improvement in ductility or oxidation resistance is obtained. Accordingly the amount of chromium is preferably maintained from about 0.3% by weight to about 5% by weight, more preferably 1-3%, to achieve a ductile material. Accordingly, in an embodiment of the invention, the higher gold gold-nickel-vanadium brazing material, with the addition of chromium, may substantially comprise, in weight percent:

- 92-98% gold
- 1.5-3.5% (preferably 1.5-2.0%) nickel
- 1-3% vanadium
- 0-1% molybdenum
- 0.3-5, preferably 1-3% chromium.

In yet further embodiments of the invention, molybdenum may be added to the gold-nickel-vanadium brazing materials, with or without chromium. The amount of molybdenum may be present in an amount up to about 4% by weight, preferably less than 1% by weight.

The following non-limitative examples illustrate the gold-nickel-vanadium brazing alloys of the present invention.

NOTE: ALL ALLOY COMPOSITIONS ARE IN WEIGHT PERCENT

Example 1

An alloy of 94% Au, 2% Ni, 1% Cr, 3% V (Alloy No. 6) was plasma melted (via a tungsten electrode) in a water-cooled copper crucible under argon gas. A 7 gram melted button of the alloy was then hot rolled down to about 75 mil [1.905mm] thickness. The alloy was subsequently rolled down to 2 mil [0.0508mm] without intermediate anneal.

A 2 mil [0.0508mm] foil was then placed between a 1 1/4" [31.75mm] x 1 1/4" [31.75mm] x 0.10" [2.54mm] alumina plate and a 1/2" [12.7mm] x 1/2" [12.7mm] x 0.040" [1.016mm] molybdenum sheet, and successfully vacuum brazed at 1,120°C under 10^{-5} Torr vacuum resulting in a brazed joint with a full fillet. The low peel strength of the joint did, however, indicate the presence of a high residual stress.

Example 2

An alloy of 96.5% gold, 1.5% nickel, 2% vanadium (Alloy No. 1) was prepared as described in Example 1 and rolled down to a 2 mil [0.0508mm] foil. Three 2 mil [0.0508mm] foils were then placed on an alumina substrate and brazed under 10^{-5} Torr vacuum at 1,050°C, 1,075°C and 1,100°C, respectively. It is possible to braze below the liquidus temperature (i.e., 1,050°C) if a wide temperature range exists between the solidus and liquidus temperature. The alloy exhibited no dewetting and melted with a bright gold colour.

The alloy was also brazed to a molybdenum test plate, as described in Example 1, and simultaneously heated to the above temperatures. The brazed joint exhibited a high peel strength, indicating a low residual joint stress.

Example 3

An alloy of 82% Au, 15.5% Ni, 0.75% molybdenum, 1.75% V (Alloy No. 14) was plasma melted (via a tungsten electrode) in a water-cooled copper crucible under argon gas. A 7 gram melted button of the alloy was then hot rolled down to about 75 mil [1.905mm] thickness. The alloy was substantially rolled down to 2 mil [0.0508mm] without intermediate anneal.

Four 2 mil [0.0508mm] foils were then placed on an alumina slab 1 1/4" [31.75mm] x 1 1/4" [31.75mm] x 0.2" [5.08mm] thick and vacuum melted under 10^{-5} Torr vacuum at temperatures of 980°C, 990°C, 1,020°C, and 1,050°C, respectively. The brazing alloy exhibited a high metallic finish and did not dewet.

Example 4

A small alumina cylinder (3/8" [9.525mm] diameter x 1/2" [12.7mm] long) was brazed to a molybdenum plate, 1" [25.4mm] x 1" [25.4mm] x 0.020" [0.508mm] thick at 1,000°C under 10^{-5} Torr vacuum, employing the alloy described in Example 3. The brazing alloy exhibited a high metallic finish and did not dewet. This joint also tested less than 10^{-9} cc/sec. on leak rate testing, using a helium mass spectrograph.

Example 5

An alloy of 82% gold, 14.5% nickel, 1% chromium, 0.75% molybdenum, 1.75% vanadium (Alloy No. 16) was prepared and tested as in Example 3 with similar excellent results.

The brazing material compositions investigated and falling within the scope of this invention (except comparative examples indicated *) are shown in Table I.

Compositions Nos. 1-6 exhibited excellent wettability to a variety of materials, including alumina ceramic. The brazed joints also exhibited a high metallic finish and a fine grain structure.

Compositions 7 and 8 are comparative examples containing titanium to be contrasted with alloys 9 and 10 respectively which contain vanadium in place of titanium. It can be seen that the vanadium containing alloys have a lower yield strength and hardness indicating a higher ductility than the comparable titanium containing alloys.

Compositions 11 & 12 are intended to compare the effect of chromium and molybdenum on ductility. It can be seen that chromium increases ductility more than the same level of molybdenum.

Compositions 14-18 exhibited excellent wettability to a variety of materials, including alumina ceramic. The brazed joints also exhibited a high metallic finish and a fine grain structure.

Compositions 19-20 also show the beneficial effect of chromium on ductility.

TABLE I

Alloy No.	ELEMENTS (% by wt.)						Solidus °C	Liquidus °C	Yield KSI	Tensile Strength KSI	Elongation %	KHN 200g
	Au	Ni	V	Ti	Cr	Mo						
1	96.50	1.50	2.00				1026	1068				
2	96.00	1.00	3.00				1060	1092				
3	94.00	2.00	4.00				1058	1091				
4	96.25	1.50	1.50		0.75		1022	1065				
5	94.00	2.00	2.00		2.00		1040	1092				
6	94.00	2.00	3.00		1.00		1041	1096				
7*	96.40	3.00		0.60			1003	1030	30.4		29	111.8
8*	97.50	0.75		1.75			1018	1031	>75.0			183.2
9	96.40	3.00	0.60				995	1030	26.4			90.5
10	97.50	0.75	1.75				987	1006	20.7			83.5
11	96.75	0.75	1.75			0.75			31.5	50.7	26.3	
12	96.75	0.75	1.75		0.75				22.6	46.8	26.6	
13	94.50	3.00	1.75			0.75			42.1	68.5	≈20	
14	82.00	15.50	1.75			0.75	950	960				190
15	82.00	15.50	1.50			1.00	950	965				
16	82.00	14.50	1.75		1.00	0.75	953	980				186
17	82.00	15.00	1.50		1.00	0.50	940	960				
18	82.00	14.25	1.00		2.00	0.75	947	968				
19	82.00	15.00	2.00		1.00							167
20	82.00	15.00	3.00									219

Units KSI are thousands of pounds per square inch and units KHN indicate Knoop hardness.

CLAIMS

1. A method of forming a brazed joint between a first material and a second material, which method comprises immediately disposing between said first and second material a brazing composition comprising in weight percent (incidental materials and volatile constituents excepted):-
 - 75-98% gold
 - 0-20% nickel
 - 0.5-6% vanadium
 - 0-5.9% molybdenum
 - 0-6% chromium.
2. A method as claimed in claim 1 in which the brazing composition comprises at least 0.25% nickel.
3. A method as claimed in claim 1 in which the brazing composition comprises in weight percent (incidental materials and volatile constituents excepted):-
 - 92-98% gold
 - 0-7.5% nickel
 - 0.5-6% vanadium
 - 0-4% molybdenum
 - 0-6% chromium.
4. A method as claimed in claim 1 in which the brazing composition comprises 0.3-3% chromium.
5. A method as claimed in claim 4 in which the brazing composition comprises 1-3% chromium.
6. A method as claimed in claim 1 in which the brazing composition comprises less than 1% molybdenum.
7. A method as claimed in claim 3 in which the brazing composition comprises 1.5-3.5% vanadium.
8. A method as claimed in claim 7 in which the brazing composition comprises 1.5-2.0% vanadium.
9. A method as claimed in claim 1 in which the brazing composition comprises less than 3% nickel.

10. A method as claimed in claim 1 in which the brazing composition comprises in weight percent (incidental materials and volatile constituents excepted):-
 - 75-92% gold
 - 0-20% nickel
 - 0.5-6% vanadium
 - 0.25-5.9% molybdenum
 - 0-6% chromium.
11. A method as claimed in claim 10 in which the brazing composition comprises 0.5-4% molybdenum.
12. A method as claimed in claim 11 in which the brazing composition comprises
 - about 82% gold
 - 14.0-16.5% nickel
 - 1-1.75% vanadium
 - 0.5-1% molybdenum
 - 0-2% chromium.
13. A brazing composition usable in the method of any of claims 1-12 and comprising an alloy foil.
14. A brazing composition usable in the method of any of claims 1-12 and comprising a paste.
15. A braze joint formable by the method of any of claims 1-12.

INTERNATIONAL SEARCH REPORT

International Application No
PCT/GB 93/01609

A. CLASSIFICATION OF SUBJECT MATTER
IPC 5 B23K35/30 C22C5/02

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 5 B23K C22C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP,A,0 005 312 (MEDTRONIC) 14 November 1979 see claims 1-3,9; examples ----	1,13,15
A	US,A,4 690 876 (H.MIZUHARA) 1 September 1987 -----	
A	US,A,4 938 922 (H.MIZUHARA) 3 July 1990 cited in the application -----	
A	DE,C,863 714 (W.C.HERAEUS G.M.B.H. PLATINSCHMELZE) 19 January 1953 -----	

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No
PCT/GB 93/01609

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP-A-0005312	14-11-79	US-A- 4180700 US-A- 4217137	25-12-79 12-08-80
US-A-4690876	01-09-87	US-A- 4606978	19-08-86
US-A-4938922	03-07-90	NONE	
DE-C-863714		NONE	